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(21) International Application Number: PCT/US98/20499 (22) International Filing Date: 30 September 1998 (30.09.98) (30) Priority Data: 9/281114 30 September 1997 (30.09.97) JP (71) Applicant (for all designated States except US): E.I. DU PONT DE NEMOURS AND COMPANY [US/US]; 1007 Market Street, Wilmington, DE 19898 (US). (72) Inventors; and (75) Inventors/Applicants (for US only): NOZAKI, Masahiro [JP/JP]; B-305 East Suzumi, 342-1, Hiramatsu Honcho, Utsunomiya, Tochigi 321-0932 (JP). KUROE, Tatsuo [JP/JP]; Park Nova Kifune, 1F, 114, Kifune 1-chome, Meito-ku, Nagoya, Aichi 465-0058 (JP). (74) Agent: HAMBY, William, H.; E.I. du Pont de Nemours and Company, Legal Patent Records Center, 1007 Market Street, Wilmington, DE 19898 (US).		(81) Designated States: CA, JP, US, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Published <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
(54) Title: POLYAMIDE COMPOSITION FOR WELDING (57) Abstract Polyamide compositions for welding are provided, having excellent weldability, heat resistance, and hydrolysis resistance. Compositions of (A) an aromatic polyamide in which the molar fraction of aromatic monomers among the monomer components making up the polyamide is at least 0.2, and (B) an aliphatic polyamide, wherein the weight ratio of (A) and (B) is from 99:1 to 5:95 are described.		

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TITLE

428 Rec'd PCT/PTO 01 MAR 2000

Polyamide Composition for Welding

Field of the Invention

- 5 The present invention relates to polyamide compositions for welding which are used, for example, in die slide injection molding and in injection molding. This invention relates more particularly to polyamide compositions for welding which contain aromatic polyamides and aliphatic polyamides.

10 Background of the Invention

- The molding of hollow molded articles from polymers such as polyamides and polyesters has traditionally relied upon blow molding and injection molding. In cases where thick-walled hollow molded articles and molded articles with a shape having thick-walled sections and thin-walled sections are produced, the
15 molded article to be produced is molded by injection molding a body that has been divided into two sections, arranging the two sections that have been removed from the molds so that they face each other at their parting lines, then heating and welding. Alternatively, the two sections that have been obtained may be placed in another mold so that they face each other at the parting lines, the molten resin
20 injected about the periphery of the opposed faces, and insert molding (or overlap molding) carried out.

- JP-B-2-38377 discloses a method for die slide injection molding, which uses a pair of molds comprising one mold provided with a male mold and a female mold for use in molding the two sections of a hollow molded article, and
25 another mold provided with a female mold and a male mold which respectively face the above male die and female die. The sections are both injection-molded at the same time using these molds, after which one of the molds is made to slide so as to cause the sections remaining in the respective female molds to become mutually opposed. By aligning the respective molds in this way, the sections are
30 mutually abutted, then molten resin is injected about the periphery of the abutted faces, thus mutually welding the sections.

- Injection welding materials which are known to be used in the production of hollow molded articles and the production of sealed molded articles include compositions including copolymers of a polyamide 6 component with a
35 polyamide 66 component, and an inorganic filler (JP-B 8-337718); compositions including resin mixtures of a polyamide 6/66 copolymer with polyamide 12, and an inorganic filler (JP-A 9-57789); and resin mixtures of polyamide 66, polyamide 12 and polyamide 6/66 copolymer, and an inorganic filler (JP-A 9-57790).

However, it will be appreciated by those skilled in the art that these materials are insufficient both with respect to weldability and heat resistance.

The object of the present invention is to provide a polyamide composition for welding which has excellent weldability, heat resistance and hydrolysis resistance. More specifically, the object of this invention is to provide polyamide compositions for molding which are highly suitable for such purposes as die sliding injection molding to produce hollow molded articles such as air intake manifolds, and injection molding to produce sealed molded components, in which an internal component has been protected from external influences, such as solenoids, aspirated air temperature sensors, and wheel speed sensors.

Summary of the Invention

There is disclosed and claimed herein polyamide compositions for welding comprising

- (A) an aromatic polyamide having a molar fraction of aromatic monomers among monomer components of said polyamide of at least 0.2, and
- (B) an aliphatic polyamide,

wherein the weight ratio of (A) and (B) is from 99:1 to 5:95.

Another embodiment of the present invention is a polyamide composition comprising aromatic polyamide (A) having a carboxylic acid component of terephthalic acid or a mixture of terephthalic acid and isophthalic acid and optionally aliphatic acid, and a diamine component of hexamethylenediamine or a mixture of hexamethylenediamine and 2-methyl pentamethylenediamine, and an aliphatic polyamide (B), wherein the weight ratio of (A) and (B) is from 99:1 to 5:95.

Yet another embodiment of the present invention is a polyamide molded article comprised of two or more members, wherein at least two of the members have been welded using the above-described polyamide composition for welding.

Brief Description of the Drawing

Figure 1 is a test piece for evaluating weldability.

Detailed Description of the Invention

The polyamide compositions of the present invention are welding compositions, and may be employed in any known welding techniques such as injection welding, vibration welding, ultrasonic welding, and spin welding.

The polyamide (A) useful in this invention is an aromatic polyamide in which the molar fraction of the aromatic monomer within the monomer components making up the polyamide is at least 0.2. In other words, this is a polyamide in which one or more of the diamine, dicarboxylic acid, and/or aminocarboxylic acid monomer components making up the polyamide is an aromatic compound, and which is composed of monomers wherein the molar fraction of aromatic monomers, based on all of the monomers, is at least 0.2.

If the molar fraction of the aromatic monomers is less than 0.2, a high heat resistance, and an excellent hydrolysis resistance and chemical resistance cannot be achieved. Moreover, properties such as these are not exhibited even in compositions. The molar fraction of the aromatic monomer is preferably from 0.25 to 0.5, and more preferably from 0.4 to 0.5.

Examples of aromatic monomers that may serve as components in the aromatic polyamide include aromatic diamines, such as p-phenylenediamine, o-phenylenediamine, m-phenylenediamine, p-xylenediamine and m-xylenediamine; aromatic dicarboxylic acids, such as terephthalic acid, isophthalic acid, phthalic acid, 2-methylterephthalic acid, and naphthalenedicarboxylic acid; and aromatic aminocarboxylic acids, such as p-aminobenzoic acid. These aromatic monomers may be used singly or as combinations of two or more thereof.

Monomers other than aromatic monomers which may be present in the aromatic polyamide include aliphatic dicarboxylic acids, aliphatic alkylenediamines, alicyclic alkylenediamines, and aliphatic aminocarboxylic acids. Exemplary aliphatic dicarboxylic acids include adipic acid, sebacic acid, azelaic acid and dodecanedioic acid; these may be used singly or in combinations of two or more thereof. Exemplary aliphatic alkylenediamine, which may be straight-chain or branched, include ethylenediamine, trimethylenediamine, tetramethylenediamine, pentamethylenediamine, hexamethylenediamine, 1,7-diaminoheptane, 1,8-diaminooctane, 1,9-diaminononane, 1,10-diaminodecane, 2-methyl pentamethylenediamine and 2-ethyl tetramethylenediamine. These aliphatic alkylenediamines may be used singly or as combinations of two or more thereof. Exemplary alicyclic alkylenediamines include 1,3-diaminocyclohexane, 1,4-diaminocyclohexane, 1,3-bis(aminomethyl)cyclohexane, bis(aminomethyl)cyclohexane, bis(4-aminocyclohexyl)methane, 4,4'-diamino-3,3'-dimethyldicyclohexylmethane, isophoronediamine and piperazine. These alicyclic alkylenediamines may be used singly or as combinations of two or more thereof. Exemplary aminocarboxylic acids include ϵ -aminocaproic acid and ω -aminoundecanoic acid, and may be used singly or as combinations of two or more thereof.

Preferable examples of the polyamide (A) include those in which the carboxylic acid component is terephthalic acid or a mixture of terephthalic acid with isophthalic acid and optionally aliphatic acid, and the diamine component is hexamethylenediamine or a mixture of hexamethylenediamine with 2-methylpentamethylenediamine.

Aliphatic polyamides (B) useful in the present invention include polyamide 66, polyamide 6, polyamide 610, polyamide 612, polyamide 46, polyamide 11, and polyamide 12, and polyamide 66 is especially preferred.

The above-described polyamides (A) and (B) are used in a weight ratio of 99:1 to 5:95, preferably 97:3 to 50:50, and more preferably 95:5 to 80:20. At an aromatic polyamide blending ratio higher than 99 or less than 5, the weldability is poor.

The polyamide composition of the present invention comprising (A) an aromatic polyamide comprising carboxylic acid component comprising aliphatic dicarboxylic acid and diamine component comprising aromatic diamine or a mixture of aromatic and aliphatic diamine, and (B) aliphatic polyamide should comprise at least 50% by weight of (A) aromatic polyamide based on a total weight of (A) and (B) polyamides. The above described polyamide (A) and (B) are used in a weight ratio of 50:50 to 95:5. If (A) polyamide is less than 50 wt. %, tensile shear strength is low and such polyamide composition cannot be used to weld polyamide molded articles composed of two or more members.

The compositions of the invention may also contain inorganic fillers in order to increase the mechanical properties. Exemplary inorganic fillers include glass fibers, carbon fibers, potassium titanate whiskers, kaolin, talc and mica, with the use of glass fibers being preferred. The amount of inorganic filler blended into the composition, based on the weight of the composition, is generally from 5 to 60% by weight, preferably from 7.5 to 50% by weight, and more preferably from 10 to 45% by weight. At less than 5% by weight, the increase in mechanical strength is insufficient, whereas blending in a large amount of more than 60% by weight results in a poor moldability.

In addition to the above-mentioned components, additives such as flame retardants, impact modifiers, heat stabilizers, plasticizers, antioxidants, nucleating agents, dyes, pigments, and parting agents may also be compounded in the inventive compositions within a range that does not compromise the properties thereof.

The polyamide compositions for welding of the present invention may be produced by a number of conventional methods as will be appreciated by those skilled in the art. Examples of such methods include a method in which two

polyamides are mixed, then are kneaded and extruded using a twin-screw extruder or some other melt-kneading apparatus, and pelletized; a method wherein two low-molecular-weight aromatic polyamides are mixed and polymerized, or a method in which extrusion polymerization is carried out concurrent with mixing.

- 5 It is also possible to carry out melt kneading and molding together using an injection molding machine.

The polyamide composition for welding of this invention is used to weld polyamide molded articles composed of two or more members. The members may each be made of a composition containing the same polyamide mixture as the
10 polyamide mixture within the polyamide composition for welding that is used, they may be made of a composition containing a mixture of the same types of polyamides but in a different blending ratio, or they may be made of a composition containing a mixture of different polyamides.

15 Examples

Examples 1-2 and Comparative Examples 1-4

The polyamide shown in Table 1 and glass fibers (chopped strands, from Nippon Sheet Glass Co., Ltd.) were melt-kneaded in a twin-screw extruder (ZSK-40, from W&P), water-cooled, then pelletized. Using the resulting pellets, the
20 primary molded article shown in Fig. 1 was injection molded. This primary molded article 1 was then set in a mold and, using the same pellets, a test piece 2 (18x100x3mm; overlapping portion 3, 18x20mm) was molded by overlap molding (secondary molding). The mold temperature and resin temperature during molding are shown in Table 1.

25 The tensile shear strength was measured using the test pieces 2 thus obtained. Measurement was carried out in accordance with ASTM D638, at room temperature, and at a test speed of 5 mm/min. The measurement results are shown in Table 2.

The heat resistance was also determined, measurement being carried out in
30 accordance with ASTM D648. The heat distortion temperature (HDT) was measured at 18.6 kg/cm². The measurement results are shown in Table 2.

In addition, the hydrolysis resistance was tested. The test piece 2 was immersed 48 hours in a 50% aqueous solution of ethylene glycol at 150°C, following which the tensile strength was measured. The tensile strength value
35 measured after 250 hours of immersion was computed as a percentage of the original measured value, and this percentage was regarded as the tensile strength retention. The results are shown in Table 2.

The polyamides in the tables were as follows.

Polymer A: A polyamide in which the dicarboxylic acid component is terephthalic acid and the diamine components are hexamethylenediamine (HMD) and 2-methyl pentamethylenediamine (MPMD) (HMD:MPMD =

5 50:50)

Polymer B: Polyamide 6

Polymer C: Polyamide 66

Polymer D: Polyamide 612

Polymer E: Polyamide 6T66 (6T:66=55:45)

10 Polymer F: Polyamide MXD6 (m-xylylenediamine-6)

Table 1

	Ex. 1	Ex. 2	Comp. Ex. 1	Comp. Ex. 2	Comp. Ex. 3	Comp. Ex. 4
Mold temperature in primary molding (°C)	120	120	120	120	120	120
Resin temperature in primary molding (°C)	310	340	290	310	290	340
Pre-heating temperature (°C)	120	120	120	120	120	120
Mold temperature in secondary molding (°C)	120	120	80	80	80	120
Resin temperature in secondary molding (°C)	310	340	290	290	290	340

Table 2

	Ex. 1	Ex. 2	Comp. Ex. 1	Comp. Ex. 2	Comp. Ex. 3	Comp. Ex. 4
Polymer A (wt %)	52.0	58.5	0	0	0	65.0
Polymer B (wt %)	0	0	70.0	0	0	0
Polymer C (wt %)	13.0	6.5	0	70.0	33.5	0
Polymer D (wt %)	0	0	0	0	33.5	0
Glass fibers (wt %)	35.0	35.0	30.0	30.0	33.0	35.0
Tensile shear strength (kg/cm ²)	232.3	200.0	217.8	84.3	102.3	88.7
Heat distortion temperature (°C)	255	260	215	254	215	260
Tensile strength retention (%)	76.6	81.0	<50	73.5	68.6	87.2

15

TABLE 3

	Ex. 3	Ex. 4	Ex. 5	Ex. 6	Ex. 7	Ex. 8
Mold temperature in primary molding (°C)	120	120	120	120	120	120
Resin temperature in primary molding (°C)	340	340	340	340	340	340
Pre-heating temperature (°C)	120	120	120	120	120	120
Mold temperature in secondary molding (°C)	120	120	120	120	120	120
Resin temperature in secondary molding (°C)	340	340	340	340	340	340

TABLE 4

	Ex. 3	Ex. 4	Ex. 5	Ex. 6	Ex. 7	Ex. 8
Polymer A (wt %)	61.75	32.5	13.0	6.50	0	0
Polymer B (wt %)	0	0	0	0	0	0
Polymer C (wt %)	3.25	32.5	52.0	58.50	80.0	20.0
Polymer D (wt %)	0	0	0	0	0	0
Polymer E (wt %)	0	0	0	0	20.0	0
Polymer F (wt %)	0	0	0	0	0	80.0
Glass fibers (wt %)	35.0	35.0	35.0	35.0	35.0	35.0
Tensile shear strength (kg/cm ²)	197.2	212.4	190.8	205.2	205.2	194.4
Heat distortion temperature (°C)	263	240	250	253	254	230
Tensile strength retention (%)	84.0	67.0	67.1	70.5	70.0	65.0

- 5 Upon comparing Examples 1 and 2 with Comparative Example 1, it is apparent that although polyamide 6 has good weldability, the heat resistance and hydrolysis resistance are insufficient.

Examples 3-8

- 10 The polyamides of Examples 3-8 and described in Tables 3 and 4 were prepared and tested in the same manner as described above.

WHAT IS CLAIMED IS:

1. A polyamide composition for welding comprising
 - (A) an aromatic polyamide having a molar fraction of aromatic monomers
5 among monomer components of said polyamide of at least 0.2, and
 - (B) an aliphatic polyamide,wherein the weight ratio of (A) and (B) is from 99:1 to 20:80.
2. A polyamide composition for welding comprising
 - 10 (A) an aromatic polyamide having a carboxylic acid component consisting of terephthalic acid, isophthalic acid or a mixture of terephthalic acid and isophthalic acid and optionally aliphatic acid; and a diamine component of aliphatic diamine, and,
 - (B) aliphatic polyamide,15 wherein the weight ratio of (A) and (B) is from 99:1 to 5:95.
3. A polyamide molded article comprised of two or more members, wherein at least two of the members have been welded using the polyamide composition for welding of Claim 1.
20
4. The composition of claim 2 wherein said aromatic polyamide comprising the carboxylic acid component being terephthalic acid or a mixture of terephthalic acid and isophthalic acid; and the diamine component being hexamethylenediamine or a mixture of hexamethylenediamine and 2-methyl
25 pentamethylenediamine.
5. A polyamide composition for welding comprising
 - (A) an aromatic polyamide wherein the carboxylic acid component is aliphatic dicarboxylic acid; and the diamine component is aromatic
30 diamine or a mixture of aromatic and aliphatic diamines, and
 - (B) aliphatic polyamide,wherein the weight ratio of (A) and (B) is from 50:50 to 95:5.

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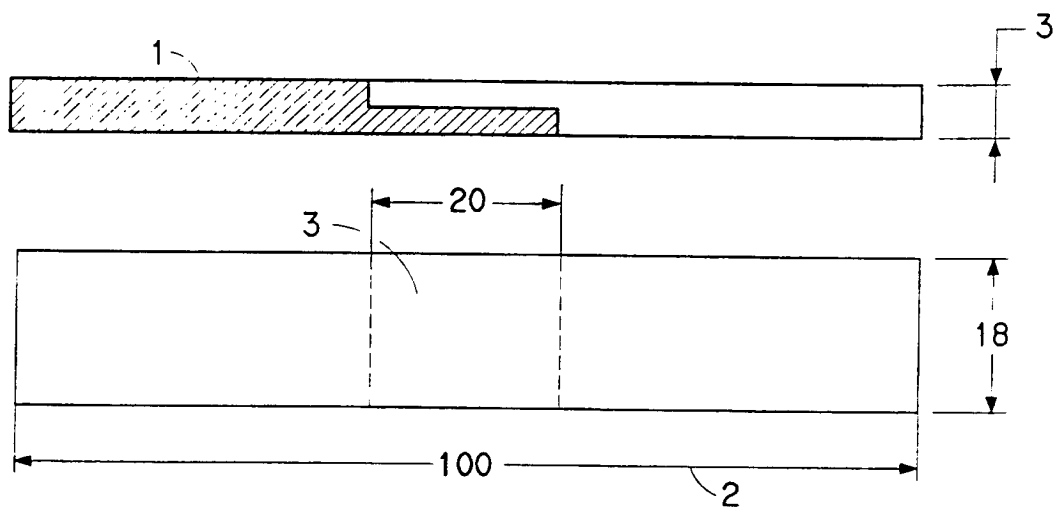
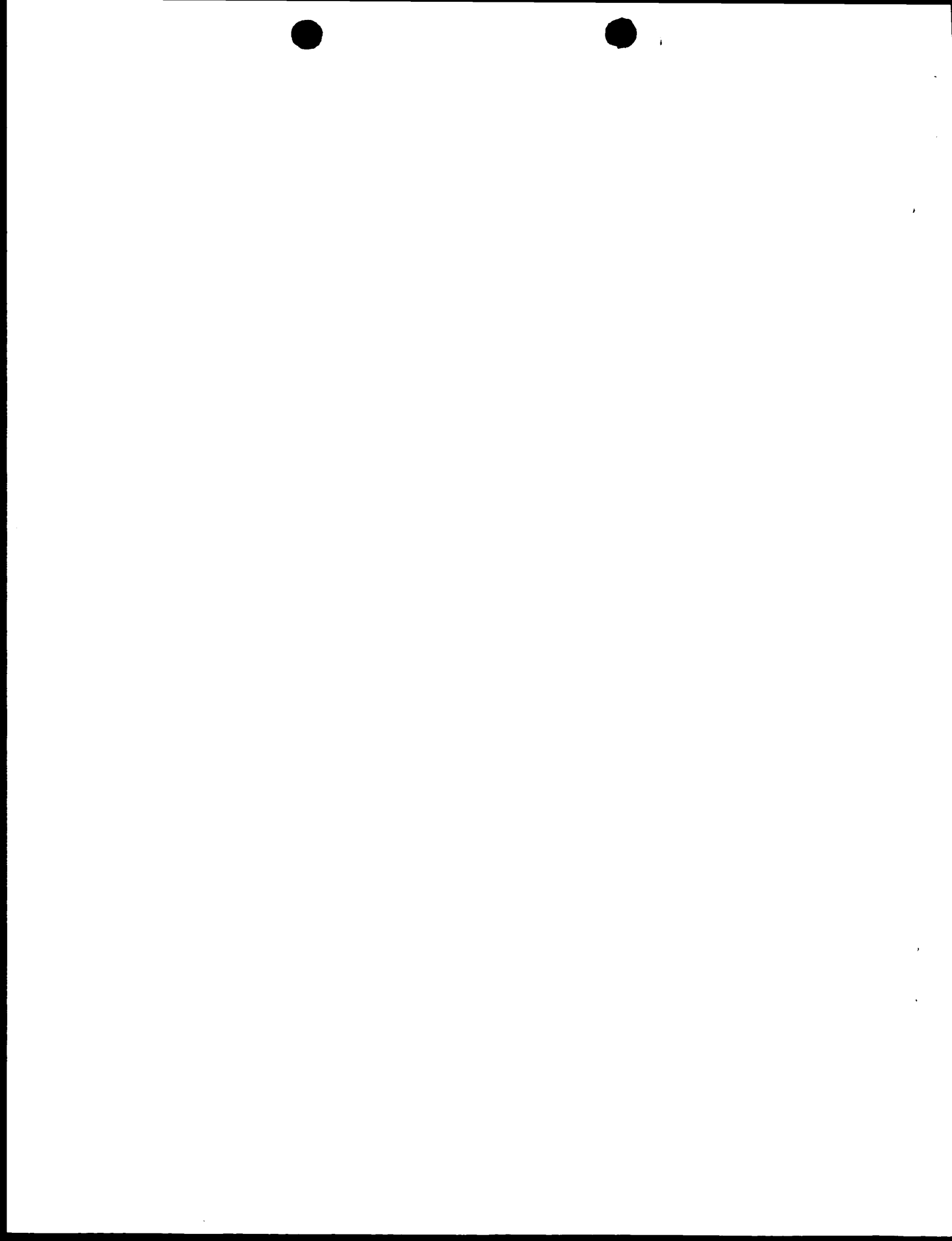


FIG. 1



INTERNATIONAL SEARCH REPORT

International Application No.

PCT/US 98/20499

A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C08L77/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No
X	EP 0 104 436 A (FELDMUEHLE AG) 4 April 1984 see page 11, line 30 - page 20, line 12; claims ---	1-4
X	EP 0 580 387 A (MITSUBISHI GAS CHEMICAL CO) 26 January 1994 see claims; examples ---	1,5
X	EP 0 458 470 A (MITSUBISHI GAS CHEMICAL CO) 27 November 1991 see claims; examples ---	1,2,4
X	EP 0 488 335 A (MITSUI PETROCHEMICAL INDUSTRIES ET AL.) 3 June 1992 see claims; examples ---	1,2,4
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Patent family members are listed in annex

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Date of the actual completion of the international search

13 January 1999

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INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication where appropriate of the relevant passages	Relevant to claim No
X	EP 0 532 963 A (BASF AG) 24 March 1993 see claims: examples ---	1,2,4
X	ELLIS T S: "COMMUNICATIONS TO THE EDITOR ON THE MISCIBILITY OF BLENDS OF NYLON 66 AND POLY(HEXAMETHYLENE ISOPHTHALAMIDE), NYLON" MACROMOLECULES, vol. 29, no. 5, 26 February 1996, pages 1836-1838, XP000555560 see the whole document ---	1,2,4
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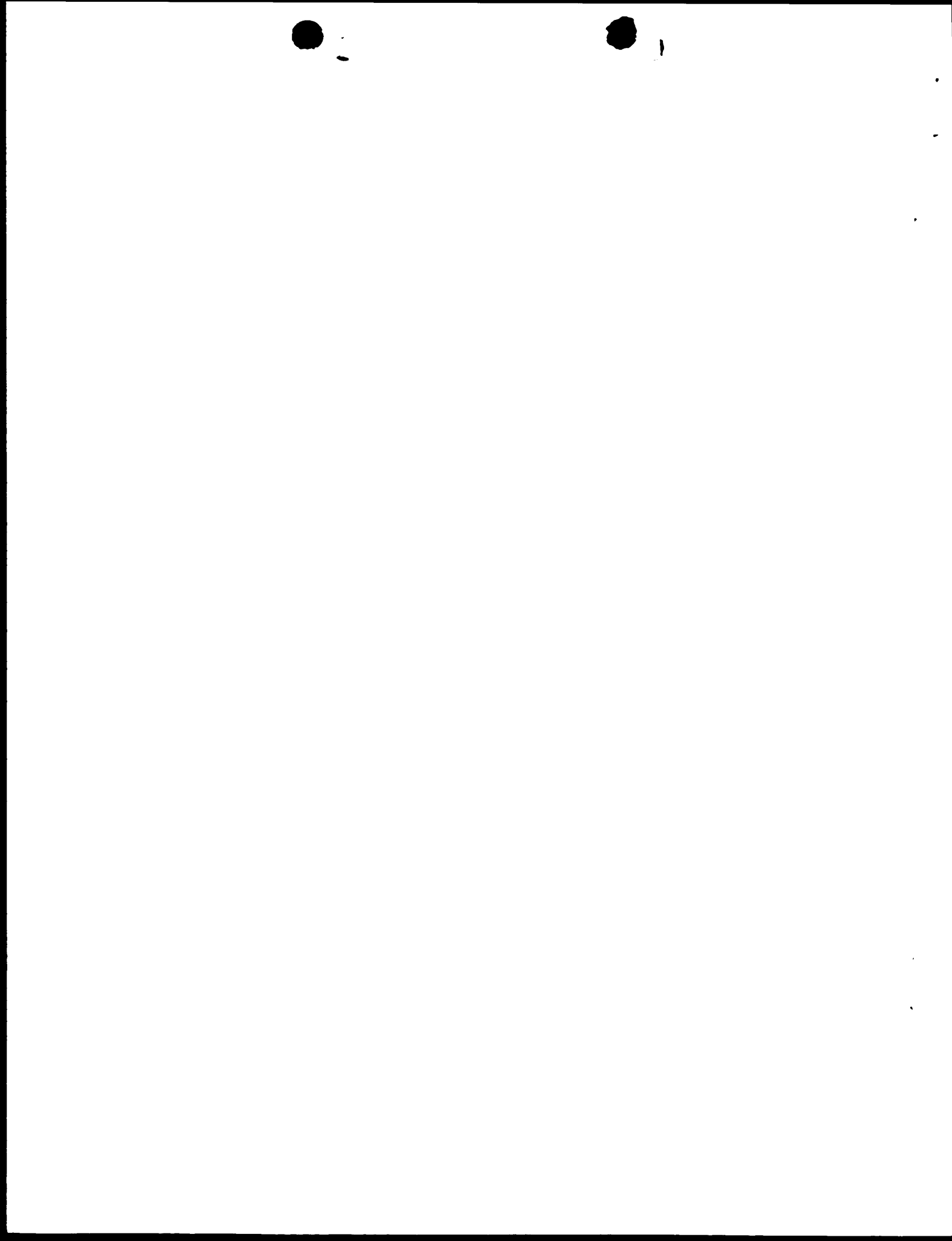
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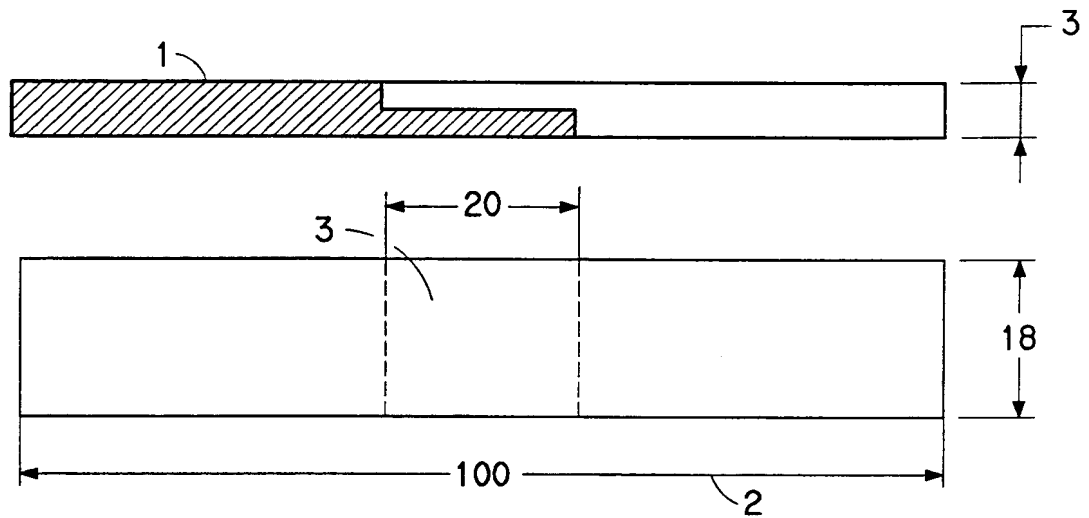


FIG. 1

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6. 1000